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PROPYLENE-TYPE RESIN-BASED MOLDED ARTICLE WITH IMPROVED CAPABILITY OF
BEING COATED AND ITS MANUFACTURING METHOD
[Hitoso-sei kairyo puropiren kei jushi seikeitai oyobi seizou houhou]

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[Claim 1] A propylene-type resin-based molded article characterized in that it comprises components (a), (b), and (c) as shown below and that the contact angle of the water droplet on the surface of said molded article is not higher than 80°;

component (a): a polypropylene-type resin containing a hydroxyl group at a concentration of at least 0.1×10^{-2} meq/g (PP) but not more than 50 meq/g(PP) in an amount of 99.9 ~ 10 weight parts,

component (b): an olefinic elastomer in an amount of 0.1 ~ 90 weight parts, and

component (c): an organic tin compound and/or tertiary amine compound in an amount of 0.01 ~ 10 weight parts per 100 weight parts of the sum of component (a) and component (b).

[Claim 2] A method of manufacturing a propylene-type resin-based molded article characterized in that a resin composition comprising components (a), (b), and (c) as shown below is heat-plasticized, supplied into a mold for resin molding with a cavity having a material with a thermal conductivity of 0.01 cal/cm·sec·°C (20 °C) or lower and a surface tension of at least 35 mN/m (20 °C) on its surface, and cooled while pressing;

component (a): a polypropylene-type resin containing a hydroxyl group at a concentration of at least 0.1×10^{-2} meq/g (PP) but not more than 50 meq/g(PP) in an amount of 99.9 ~ 10 weight parts,

* Claim and paragraph numbers correspond to those in the foreign text.

component (b): an olefinic elastomer in an amount of 0.1 ~ 90 weight parts, and

component (c): an organic tin compound and/or tertiary amine compound in an amount of 0.01 ~ 10 weight parts per 100 weight parts of the sum of component (a) and component (b).

[Detailed explanation of the invention]

[0001] [Industrial application areas]

The present invention relates to a propylene-type resin-based molded article exhibiting excellent capability of being coated with a urethane-type coating. The propylene-type resin-based molded article of the present invention can be preferably used in areas of automotive parts, electric parts, machine parts, toys, stationary products, articles for daily usage and the like, especially in areas in which coating/adhesion is necessary.

[0002] [Prior arts]

Because a propylene-type resin has balanced characteristics of good moldability, low cost, good mechanical properties, good chemical resistance, and good electric properties, it has been widely used in the areas of automotive parts, electric/electronics parts, machine parts and the like. However, since the propylene-type resin does not have a polar group, it exhibits difficulty in being coated and in obtaining a coated film with a sufficient adhesive strength, which has been an obstacle in exploring a new application area for the propylene-type resin.

[0003] As methods of improving capability of being coated and adhesion of the coated film of the propylene-type resin, a dry surface treatment method such as a flame treatment, a plasma treatment, an ozone treatment, a corona-discharge treatment, and a UV or electron beam treatment and the like, a wet surface treatment method such as using a mineral acid such as a mixed chromic acid solution, concentrated sulfuric acid and the like, chemically grafting a surface modifier component on the surface of a molded article and the like, or a primer coating method in which a surface modifier layer is formed on the surface of a molded article by direct coating, can be mentioned. However, the above-mentioned methods have not been suitable methods in terms of required equipment and productivity.

[0004] On the other hand, there has been a method of improving its capability of being coated by grafting a polar group-containing compound to a propylene-type resin-based molded article by using a peroxide compound. This method has recently attracted special attention because this method has an economical advantage in terms of simple treatment equipment and a simple treatment process. However, this method has a disadvantage in that as the grafting reaction proceeds, the chain scission reaction of the propylene-type resin proceeds at the same time, causing deterioration in mechanical properties of the resin, such as stiffness, impact resistance, heat resistance and the like, which has sometimes limited the application areas of the resin. Also, when the degree of grafting is increased to obtain sufficient adhesive strength, problems such as increased amount of bleeding

materials, generation of offensive odor, color change and the like, have been occasionally observed.

[0005] [Problems to be Solved by the Invention]

The purpose of the present invention is to provide a propylene-type resin-based molded article which has an excellent capability of being coated while maintaining balanced characteristics of moldability, cost, mechanical properties, chemical resistance, electrical properties and the like.

[0006] [Means to Solve the Problem]

The present inventors have investigated to obtain a propylene-based resin-based molded article which exhibits excellent capability of being coated and excellent adhesive strength of the coated film and discovered that there is a correlation between the contact angle of the water droplet on the surface of the above molded article and the adhesion of the coated film. That is, it has been discovered that a molded article based on a polypropylene-type resin having a specific concentration range of hydroxyl groups in a polypropylene molecule exhibits much improved adhesion of the coated film when the contact angle of the water droplet on its surface is not higher than 80°. Furthermore, it has been discovered that when the propylene-type resin is molded using a mold having a specific surface, the orientation of the polar group to the surface of the molded article can be effectively promoted, driven by the interlayer affinity between the polar group and the mold surface. In this method, the propylene-type resin-based molded article exhibiting excellent adhesion of the coated

film can be obtained without sacrificing the easy molding process and high productivity of the propylene-type resin. Furthermore, since this molded article of the present invention exhibits excellent capability of being coated even at a low degree of grafting of the polar group-containing compound, problems associated with the above chemical modification, such as deterioration in the physical properties, increased bleeding compounds, generation of offensive odor, color change and the like, can be prevented.

[0007] That is, the propylene-type resin-based molded article of the present invention is characterized in that it comprises components (a), (b), and (c) as shown below and that the contact angle of the water droplet on the surface of the above molded article is not higher than 80°;

component (a): a polypropylene-type resin containing a hydroxyl group at a concentration of at least 0.1×10^{-2} meq/g (PP) but not more than 50 meq/g(PP) in an amount of 99.9 ~ 10 weight parts,

component (b): an olefinic elastomer in an amount of 0.1 ~ 90 weight parts, and

component (c): an organic tin compound and/or tertiary amine compound in an amount of 0.01 ~ 10 weight parts per 100 weight parts of the sum of component (a) and component (b).

As methods of obtaining the molded article having the above-mentioned surface; a resin composition having the above-mentioned components is supplied to a mold for resin molding having a material with a surface tension of at least 35 mN/m (20 °C) on its cavity surface and is melt-pressed; or a heat-plasticized above resin is supplied to the mold for resin molding

having a material with a surface tension of at least 35 mN (20 °C) on its cavity surface which is pre-heated to at least 100 °C and cooled while pressing, can be mentioned.

[0008] Furthermore, the molded article having the above-mentioned surface property can be obtained by supplying the heat-plasticized resin having the above-mentioned composition into a mold for resin molding having a material with a thermal conductivity of 0.01 cal/cm·sec·°C (20 °C) or lower and a surface tension of at least 35 mN/m (20 °C) on its cavity surface, and cooling while pressing the above resin composition. In this method, a surface with excellent capability of being coated can be obtained without losing the easy moldability and high productivity of the resin.

[0009] The present invention will be explained in detail below.

(1) The resin composition

The molded article of the present invention comprises the following components (a), (b) and (c).

Component (a): a polypropylene-type resin containing a hydroxyl group at a concentration of at least 0.1×10^{-2} meq/g (PP) but not more than 50 meq/g (PP) in a amount of 99.9 ~ 10 weight parts;

component (b): an olefinic elastomer in an amount of 0.1 ~ 90 weight parts; and

component (c): an organic tin compound and/or a tertiary amine in an amount of 0.01 ~ 10 weight parts per 100 weight parts of the sum of components (a) and (b).

To obtain the propylene-type resin-based molded article exhibiting strong adhesive strength to the coated film, the component ratio of (a) to (b) is preferably in the range of 99 ~ 10 weight parts of component (a) to 1 ~ 90 weight parts of component (b), or more preferably 90 ~ 30 weight parts of component (a) to 10 ~ 70 weight parts of component (b). Also, the tin compound and/or the tertiary amine compound are used in an amount of preferably 0.05 ~ 5 weight parts per the sum of component (a) and component (b), or more preferably 0.1 ~ 3 weight parts.

[0010] Below, the components constituting the resin composition of the present invention will be explained in detail.

(a) Polypropylene-type resin containing the hydroxyl group

The polypropylene-type resin containing the hydroxyl group which constitutes the resin composition to be used in the present invention can be obtained by reacting polypropylene grafted with at least one kind of unsaturated carboxylic acid or unsaturated dicarboxylic acid anhydride (hereinafter referred to as acid-modified polypropylene) with an organic compound containing at least one primary amino group and at least one hydroxyl group in a molecule (hereinafter referred to as a hydroxyl group-containing amine compound). The concentration of the hydroxyl group is at least 0.1×10^{-2} meq/g (PP) but not more than 50 meq/g (PP). When the concentration of the hydroxyl group is in the above range, the obtained modified polypropylene can be used together with an unmodified polypropylene. The concentration of the hydroxyl group means the concentration of the hydroxyl group introduced to the polypropylene

molecule by the chemical bonding, not including the hydroxyl group attached to the residual unreacted amine compound present in the resin composition. When the concentration of the hydroxyl group is less than 0.1×10^{-2} meq/g (PP), the obtained resin-molded article does not exhibit the effect of improvement in the capability of being coated. When the concentration exceeds 50 meq/g (PP), deterioration in the mechanical strength of the molded article and a generation of a large quantity of bleeding materials on the surface of the molded article are observed, which is not preferable. The optimum concentration of the hydroxyl group in the resin is $0.5 \times 10^{-2} \sim 10$ meq/g (PP), or more preferably $1 \times 10^{-2} \sim 5$ meq/g (PP).

[0011] Below, a method of manufacturing the hydroxyl group-containing polypropylene-type resin using the acid-modified polypropylene as a starting material will be explained in more detail.

(a-1) Acid-modified polypropylene

The acid-modified polypropylene to be used in the manufacture of the hydroxyl group-containing polypropylene can be obtained by a conventional manufacturing method in which polypropylene is grafted with an unsaturated carboxylic acid or an unsaturated dicarboxylic acid anhydride.

[0012] The concentration of the unsaturated carboxylic acid group or the unsaturated dicarboxylic acid anhydride group in the acid-modified polypropylene is in the range of 0.01 ~ 10 weight%, or more preferably 0.1 ~ 5 weight%. When the concentration of the unsaturated carboxylic acid group or the unsaturated dicarboxylic acid anhydride group is less than 0.01 weight%, the capability of being coated and the adhesive strength of the

coated film, which are the purpose of the present invention, cannot be achieved. On the other hand, when the concentration exceeds 10 weight%, the final resin composition exhibits deteriorated mechanical strength, which is not preferable.

[0013] The polypropylene to be used in manufacture of the acid-modified polypropylene is propylene homopolymer or a copolymer of propylene with other α -olefins. As examples of α -olefins to be copolymerized with propylene, ethylene, butane-1, pentene-1, 2-methylbutene-1, 3-methylbutene-1, hexane-1, 3-methylpentene-1, 4-methylpentene-1, 3,3-dimethylbutene-1, heptene-1, methylhexene-1, dimethylpentene-1, trimethylbutene-1, ethylpentene-1, octane-1, methylpentene-1, dimethylhexene-1, trimethylpentene-1, ethylhexene-1, methylethylpentene-1, diethylbutene-1, propylpentene-1, decene-1, methylnonene-1, dimethyloctene, trimethylheptene-1, ethyloctene-1, methylethylheptene-1, diethylhexene-1, dodecene-1, hexadodecene and the like, can be mentioned. Those copolymers of α -olefin and propylene can be a random copolymer or an α -olefin-propylene block copolymer. Also, those α -olefins can be used alone or as a mixture of two or more kinds. Also, it is preferable that the amount of α -olefin in the in the polypropylene is not more than 45 mol%. Among those polypropylenes, propylene homopolymer, crystalline propylene-ethylene block copolymer with an ethylene content of 2 ~ 40 mol% and crystalline propylene-ethylene random copolymer with an ethylene content of 0.5 ~ 10 mol% are preferably used.

Those polypropylenes preferably have a melt index of 0.05 ~ 100 g/10

minutes, or more preferably 0.1 ~ 40 g/10 minutes when measured at 230 °C and 2.16 kg load for better moldability. Those polypropylenes can be used alone or as a combination of two or more kinds. Those polypropylenes can be manufactured in a variety of methods. For example, they can be synthesized in the presence of a combination of a solid titanium catalyst component and an organic metal catalyst component.

[0014] Also, as examples of unsaturated carboxylic acids or unsaturated dicarboxylic acid anhydrides to be grafted on the acid-modified [sic] polypropylene, carboxyl group-containing unsaturated compounds such as acrylic acid, methacrylic acid, crotonic acid, cinnamic acid, itaconic acid and the like, dicarboxylic acids such as maleic acid, fumaric acid, itaconic acid, chloromaleic acid, citraconic acid, allysuccinic acid, mesaconic acid, aconitic acid and the like and their anhydrides, can be mentioned. Among those, maleic anhydride, acrylic acid, methacrylic acid are preferably used. Especially, maleic anhydride is the most preferable. Those monomers can be used alone or in combination of two or more kinds. Also, as described in Patent Publication Hei-236214, by reacting the above compounds together with an unsaturated aromatic monomer at a specified mixing ratio, the amount of the addition of the unsaturated carboxylic acid or unsaturated dicarboxylic acid anhydride to the polypropylene can be increased.

[0015] To manufacture the acid-modified polypropylene, conventional methods can be used. That is, a method of first heat-dissolving polypropylene in an organic solvent, then reacting with the unsaturated

carboxylic acid or the unsaturated dicarboxylic acid anhydride in the presence of a radical-generating agent (solution method), a method of melting polypropylene by heating at a temperature higher than the melting point, then reacting it with the unsaturated carboxylic acid or the unsaturated dicarboxylic acid anhydride in the presence of a radical-generating agent (melting method), and a method of radiation grafting using an electron beam and the like, can be mentioned.

[0016] In the solution method, it is preferable to use an aromatic solvent, such as xylene. The reaction temperature is in the range of 100 ~ 180 °C. This method has characteristics in that this reaction proceeds with less side reaction and that the modified polypropylene having uniformly distributed unsaturated carboxylic acid groups or unsaturated dicarboxylic acid anhydride groups can be obtained. On the other hand, in the melt method, the starting material resin is reacted at a temperature of the melting point of the resin or higher, but not higher than 300 °C using a banbury mixer, kneader, or a single or a multi-screwed extruder. In the melt method, the operation is simple and the reaction can be completed in a short time. In the kneading, it is preferable that resin components in a form of a powder or pellet are previously uniformly mixed in a tumbler or a Henschel mixer. However, if necessary, the above mixing process can be omitted and each component can be separately charged into a kneading apparatus at specified amounts.

[0017] Although the radical-generating agent to be used in the reaction can be appropriately selected from conventionally known compounds,

organic peroxides are especially preferable. As preferable examples, benzoyl peroxide, dicumyl peroxide, lauroyl peroxide, 2,5-dimethyl-2,5-di-(t-butylperoxy)hexane, 2,5-dimethyl-2,5-di-(t-butylperoxy)hexyne-3, 1,3-bis(t-butylperoxyisopropyl)benzene, 1,1-bis(t-butylperoxy)-3,3,5-trimethylcyclohexane, n-butyl-4,4'-bis(t-butylperoxy)valerate, p-chlorobenzoyl peroxide, 2,4-dichlorobenzoyl peroxide, t-butylperoxyisopropyl carbonate, di-t-butyl peroxide, t-butyl peroxybenzoate, cumene hydroperoxide, diacetyl peroxide, lauroyl peroxide, t-butyl cumylperoxide and the like, can be mentioned.

[0018] Also, unreacted components (unsaturated carboxylic acid or unsaturated dicarboxylic acid anhydride, radical-generating agent), its oligomer, and byproducts such as decomposed compounds can be removed by the suction action of a vacuum pump through a vent line which is attached at the middle zone or in the vicinity of the exit of the extruder or by dissolving the reaction product in an appropriate solvent and precipitation.

[0019] Furthermore, the obtained acid-modified polypropylene can be heat-treated at a temperature of 60 °C or higher or can be subjected to vacuum suction under the molten state to remove unreacted components, oligomer, byproducts such as decomposed compounds and the like.

(a-2) Manufacturing method of hydroxyl group-containing polypropylene-type resin.

Conventional methods can be used to manufacture a hydroxyl group-containing polypropylene-type resin from the above acid-modified polypropylene. That is, a method of reacting the acid-modified polypropylene with the hydroxyl group-containing amine compound by heat-dissolving above reactants in an organic solvent (solution method) or a method of reacting the above reactants after heat-melting the reactants at a temperature of melting points of the reactants or higher (melt method) can be used.

[0020] As favorable examples of hydroxyl group-containing amine compounds to be used in manufacture of the hydroxyl group-containing polypropylene-type resin, 2-aminoethanol, 3-amino-1-propanol, 4-amino-1-butanol, 5-amino-1-pentanol, 6-amino-1-hexanol, 2-amino-1-butanol, 2-(2-aminoethoxy)ethanol, N-aminoethyl ethanolamine, 2-amino-3-methyl-1-butanol, 2-amino-2-methyl-1-propanol, 2-amino-1-propanol, 3-amino-1,2-propanediol, 3-amino-1,3-propanediol, 2-amino-2-methyl-1,3-propanediol, 2-(3-aminopropylamino)ethanol, 2-amino-2-ethyl-1,3-propanediol, tris(hydroxymethyl)aminomethane, o-, p-, m-aminophenol, 2-aminophenetyl alcohol, 4-aminophenetyl alcohol, o-, p-, m-aminobenzyl alcohol, 2-amino-4-methylphenol, 2-amino-5-methylphenol, 5-amino-2-methylphenol, 2-amino-4-chlorophenol, 4-amino-2,6-dichlorophenol and the like, can be mentioned. Also, a partially aminated aliphatic polyhydric alcohol derived from polyethylene

glycol, polypropylene glycol, or a triol can be used. When the partially aminated aliphatic polyhydric alcohol is to be used, that with a number-average molecular weight of 10,000 or less is preferably used, or more preferably 100 ~ 2,000. Among those compounds, 2-aminoethanol, 3-amino-1-propanol, 4-amino-1-butanol, 5-amino-1-pentanol, 6-amino-1-hexanol, and 2-(2-aminoethoxy)ethanol are especially preferable.

[0021] In the manufacture of the hydroxyl group-containing polypropylene-type resin, the above-mentioned hydroxyl group-containing amine compounds can be used alone or as a combination of two or more kinds.

(b) Olefinic elastomer

The olefinic elastomer constituting the resin composition of the present invention is a copolymer containing α -olefins, such as ethylene, propylene, butane-1, pentene-1 and the like, or a copolymer of those α -olefins with an nonconjugated diene. As examples of nonconjugated dienes, dicyclopentadiene, 1,4-hexadiene, dicyclooctadiene, methylenenorbornene, 5-ethylidene-2-norbornene and the like, can be mentioned.

[0022] As concrete examples of olefinic elastomers, amorphous elastomer copolymers mainly consisting of an olefin, such as ethylene-propylene copolymer rubber, ethylene-butene-1 copolymer rubber, ethylene-propylene-butene-1 copolymer rubber, ethylene-propylene-nonconjugated diene copolymer rubber, ethylene-butene-1-nonconjugated diene copolymer rubber, and ethylene-propylene-butene-1-nonconjugated diene copolymer rubber can be

mentioned. Those elastomers can be used alone, or in combination of two or more kinds.

[0023] Also, the above olefinic elastomers have Mooney viscosity ML_{1+4} (100 °C) of 5 ~ 150, or more preferably 10 ~ 120. The olefinic elastomers preferably have an iodine number (degree of unsaturation) of 16 or less.

(c) Organic tin compound and/or tertiary amine compound.

The tin compound which constitutes the resin composition of the present invention has a formula as shown below.

[0024] [Chemical structure 1]



(In the above formula, R_1 is an alkyl group with carbon atom numbers of 4 ~ 10; X_1 is an alkyl group with carbon atom numbers of 4 ~ 10, chlorine atom, or a hydroxyl group; and Y_1 and Y_2 can be the same or different and are chlorine atom, $-OCOR_2-$, or a hydroxyl group, with R_2 being an alkyl group, an aryl group, or allylalkyl group.)

[0025] As concrete examples of these tin compounds, $n-C_4H_9Sn(OH)_2Cl$, $n-C_4H_9Sn(OH)Cl_2$, $n-C_4H_9SnCl_3$, $C_8H_{17}Sn(OH)_2Cl$, $C_8H_{17}Sn(OH)Cl_2$, $C_8H_{17}SnCl_3$, $n-C_4H_9Sn(OH)_2OCOC_7H_{15}$, $n-C_4H_9Sn(OH)_2OCOC_{11}H_{23}$, $n-C_8H_{17}Sn(OH)_2OCOC_7H_{15}$, $n-C_8H_{17}Sn(OH)_2OCOC_{11}H_{23}$, $n-C_4H_9Sn(OCOC_7H_{15})_3$, $(n-C_4H_9)_2Sn(OCOC_{11}H_{23})_2$, $(n-C_8H_{17})_2Sn(OCOC_{11}H_{23})_2$, $(n-C_4H_9)_2Sn(OCOCH=CHCOOCH_3)_2$, $(n-C_4H_9)_2Sn(OCOCH=CHCOOCH_2Ph)_2$ and the like, can be used. Among those, $n-C_4H_9SnCl_3$, $(n-C_4H_9)_2Sn(OCOC_{11}H_{23})_2$, and $(n-C_8H_{17})_2Sn(OCOC_{11}H_{23})_2$ are preferably used.

[0026] Also, as examples of tertiary amine compounds which constitutes the resin composition of the present invention, dimethylpropylamine, diethylpropylamine, tris(dimethylaminomethyl)phenol, tetraguanidine, N,N-dibutylethanolamine, N-methyl-N,N-diethanolamine, 1,4-diazabicyclo[2,2,2]octane, 1,8-diazabicyclo[5,4,0]-7-undecene, tetramethylbutanediamine and the like, can be mentioned.

[0027] In the resin composition to be used in the present invention, the above-mentioned tin compounds and tertiary amine compounds can be used alone or in combination of two kinds or more, including a combination of the tin compound and the tertiary amine compound.

Manufacturing method of the resin composition

To manufacture the propylene-type resin composition to be used in the present invention, components (a), (b), and (c) are mixed in a variety of methods. There is no restriction in the order of mixing. Components (a), (b), and (c) can be mixed all at once, or component (c) can be mixed with a mixture of components (a) and (b), or component (b) can be mixed with a mixture of components (a) and (c), or component (a) can be mixed with a mixture of components (b) and (c).

[0028] There are a variety of methods in manufacture of the propylene-type resin composition to be used in the present invention. For example, components (a), (b), and (c) are uniformly pre-mixed using equipment such as a tumbler or a Henschel mixer and the resin composition is formed inside an extruder or injection molding equipment during the

molding process in manufacture of a film or a molded article. Or, a method of mixing the components (a), (b) and (c) at a temperature of melting point of the raw material resins or higher but not higher than 300 °C using a banbury mixer, a kneader, an extruder and the like (melt kneading method) can be used. Or, components (a) and (b) are first pre-mixed, then component (c) can be added during the molding process.

[0029] Among the above-mentioned manufacturing methods, the melt kneading method has an advantage in that this process requires a simple operation and the resin composition can be palletized in a short time. In this melt kneading process, it is preferable that the components are uniformly pre-mixed using equipment such as a tumbler or a Henschel mixer. However, the above process can be omitted if necessary and each component can be separately charged at a specified amount into the kneading equipment. As the kneading equipment, a multi-screw extruder is preferably used because the reaction [sic] proceeds effectively using this equipment.

[0030] When an extruder is to be used, the propylene-type resin composition can be manufactured in an one-step extrusion process by first reacting polypropylene with the unsaturated carboxylic acid or its anhydride in the presence of a radical-generating agent at the initial zone of the extruder to form an acid-modified polypropylene, then adding the hydroxyl group-containing amine compound at the middle or later zone of the extruder, and further supplying components (b) and (c) to the extruder. This method is quite effective in reducing the manufacturing cost.

[0031] Also, the propylene-type resin composition to be used in the present invention can contain, as necessary, additives such as reinforcing agents, fillers, colorants (pigment, dye), UV absorbers, heat stabilizers, flame retardants, antioxidants, antistatic agents, antifogging agents, lubricants, blowing agents, plasticizers and the like, during the manufacturing process or later during the molding process in an amount which will not negatively affect the physical properties of the resin. Furthermore, the propylene-type resin composition to be used in the present invention can be mixed with other thermoplastic resins which are highly compatible with the propylene-type resin composition depending on the usage and the purpose. As examples of such resins, homopolymers such as linear low-density polyethylene (LLDP), low-density polyethylene (LDPE), very-low-density polyethylene (VLDPE), high-density polyethylene (HDPE), polybutene, polyisobutene, poly(4-methyl-1-pentene) and the like, a variety of ethylene copolymers such as ethylene-vinyl acetate copolymer (EVA), ethylene-vinyl alcohol copolymer, (EVOH), ethylene-maleic anhydride copolymer, ethylene-unsaturated carboxylic acid copolymer and ethylene-alkyl (meth)acrylate copolymer, styrene-butadiene-(styrene) block copolymer and its hydrogenated copolymer, styrene-isobutylene-(styrene) block copolymer and its hydrogenated copolymer and the like, can be mentioned. Those polymers can be used alone or as a combination of two or more kinds. Also, the above-mentioned copolymers include random, block, random-block, and graft copolymers. The number-average molecular weights of those thermoplastic resins are

normally in the range of 500 ~ 500,000, or preferably in the range of 1,000 ~ 50,000. Also, those thermoplastic resins can be used in an amount of 0 ~ 500 weight parts per 100 weight parts of the propylene-type resin composition to be used in the present invention.

As concrete examples of reinforcing agents and fillers to be used in the propylene-type resin composition of the present invention, reinforcing fibers such as glass fiber, asbestos fiber, carbon fiber, silica fiber, silica-alumina fiber, alumina fiber, zirconia fiber, boron nitride fiber, silicon nitride fiber, boron fiber, and the like, and inorganic fillers such as fumed silica, clay (aluminum silicate), glass bead, carbon black, quartz powder, talc (magnesium silicate), titanium oxide, iron oxide, calcium carbonate, diatomaceous earth and the like, can be mentioned.

[0032] For fibers, those having an average fiber diameter of 5 ~ 30 μm and a fiber length of 30 ~ 50 μm can be used. Especially for the glass fiber, that which is surface-treated with a variety of coupling agents to improve interfacial adhesion with the propylene-type resin composition and dispersibility can be used. Examples of coupling agents include conventional silane-type and titanium-type coupling agents. Although untreated inorganic fillers can be used, those which are treated with a variety of silane coupling agents and surfactants such higher fatty acids, higher fatty acid esters, higher fatty acid amides, higher fatty acid salts and the like, can be used to improve the interfacial adhesion with the propylene-type resin composition and to improve dispersibility. The

inorganic filler preferably has an average particle diameter of not more than 5.0 μm , or more preferably has an average diameter of not more than 5.0 μm and an aspect ratio of at least 5. Among above inorganic fillers, talc is especially favorably used.

[0033] The amount of the reinforcing agent and/or the filler to be used is 0 ~ 40 weight parts per 100 weight parts of the propylene-type resin composition. When the amount of the reinforcing agent and/or the filler to be used exceeds 40 weight parts, the impact resistance of the resin composition drastically deteriorates, which is not preferable. They are preferably incorporated in an amount of 5 ~ 30 weight parts. Those reinforcing agents and fillers can be used alone or as a combination of two or more kinds. When the inorganic filler and the glass fiber are to be used together, the ratio of the inorganic filler to the glass fiber is preferably 20 ~ 80 weight% of the inorganic filler to 80 ~ 20 weight% of the glass fiber.

(2) Molding method

The method of molding to obtain the molded article of the present invention will be described below. The molded article of the present invention can be obtained by orienting the polar group in the resin composition at the surface of the molded article which is driven by the affinity of the polar group (hydroxyl group) and the surface of the mold for resin molding.

[0034] The contact angle of the water droplet at the surface of the molded article can be a measure of the relative degree of the surface

orientation of the polar group at the surface. As the surface orientation of the polar group progresses, the surface becomes hydrophilic and the contact angle of the water droplet becomes lower. The molded article based on the propylene-type resin composition of the present invention, which exhibits excellent capability of being coated, has a water contact angle of 80° or lower, or preferably 75° or lower. In general, the lower contact angle indicates the higher degree of progress in the surface orientation of the polar group and better capability of being coated. However, in the resin composition of the present invention, obtaining a contact angle of lower than 60° is practically difficult.

[0035] There is no restriction in the method of obtaining the molded article of the present invention as long as it can effectively promote the surface orientation of the polar group. As concrete examples, the following methods can be mentioned: (1) a method of supplying the resin composition to a mold for resin molding with a cavity having a material with a surface tension of at least 35 mN/m (20°C) on its surface and melt-pressing the resin; (2) a method of pre-heating a mold for resin molding with a cavity having a material with a surface tension of at least 35 mN/m (20°C) on its surface to a temperature at 100 °C or higher utilizing high-frequency heating equipment, supplying the heat-plasticized resin into the above mold, and cooling the resin while pressing; and (3) a method of supplying a heat-plasticized resin composition into a mold for a resin molding with a cavity having a material with a thermal conductivity of not higher than 0.01 cal/cm·sec·°C (20°C) and a surface tension of at least

35 mN/m (20°C) on its surface and cooling the resin while pressing.

[0036] Among the above-mentioned methods, method (3) is quite preferable because a surface of the molded article having a capability of being coated can be obtained with a minimum sacrifice of the ease of molding process and the productivity. Below, method (3) will be explained in more detail. The mold for a resin molding with a cavity having a material with a thermal conductivity of 0.01 cal/cm·sec·°C (20°C) or lower and a surface tension of at least 35 mN/m (20°C) on its surface can be obtained by coating a material satisfying the above conditions onto the cavity surface of the mold. When the material to be used on the mold surface has a thermal conductivity of higher than 0.01 cal/cm·sec·°C, a rapid solidification of the molten resin proceeds. In this case, orientation of the polar groups in the resin to the surface side does not proceed effectively, resulting in the insufficient improvement in the capability of being coated and the adhesion property. Even when a material having a thermal conductivity of higher than 0.01 cal/cm·sec·°C is used, the effect of improvement can be observed by setting the higher mold temperature [as a measure, a temperature higher than (melting point-50°C)]. However, this causes a longer solidification time, resulting in decreased efficiency of molding, which is not preferable. When the thermal conductivity becomes lower, a heat-insulating effect becomes larger, which is preferable. However, in general, a minimum obtainable practical thermal conductivity is 10^{-5} cal/cm·sec·°C.

[0037] It is necessary that the material to be used on a mold surface has a surface tension of at least 35 mN/m at 20 °C measured by the Zisman method. When the surface tension is lower than 35 mN/m, it becomes difficult to orient the polar group to the surface of the molded article during the molding. A higher surface tension is preferable. However, it is generally lower than 60 mN/m, taking into consideration the balance with the thermal conductivity.

[0038] As preferable materials to be used on the mold surface which satisfy the above conditions in manufacture of the molded article of the present invention, resins, ceramics, resin/ceramic composite materials, resin/metal composite materials, resin/metal oxide composite materials, ceramic composite materials, and the like having a surface tension of at least 35 mN/m at 20 °C, can be mentioned. Also, after coating the mold with the material to be used on a mold surface, the surface of the above material can be subjected to a secondary treatment, such as physical or chemical treatment to control the surface tension.

[0039] Among above-mentioned materials to be used on the mold surface, resins and resin-type composite materials are especially preferable since they have thermal conductivities in the order of 10^{-4} (cal/cm·sec·°C), giving a large heat-insulating effect. Since a ceramic-type material has a slightly higher thermal conductivity than the resin material on the order of 10^3 [sic], the coating thickness of the ceramic-type material has to be slightly higher than that of the resin material to obtain a similar level of heat-insulating effect.

[0040] The above-mentioned material to be used on a mold surface preferably has properties of, other than the above-mentioned conditions, excellent heat resistance, excellent resistance to cooling/heating cycle, excellent abrasion resistance, excellent coatability to the mold, excellent adhesion to the mold, capability of being subjected to surface polishing, and the like. The material to be used on the mold surface which is practically coated on the outermost layer of the mold surface preferably has a low thickness in terms of preventing an increase in the cooling time. The optimum thickness of the coated film of the material to be used on a mold surface to obtain a sufficient surface modification effect without negatively affecting the molding efficiency varies depending of the thermal conductivity of the material to be used, the resin temperature during molding, the mold temperature, the injection rate of the resin, and the like. Therefore, the film thickness of the heat-insulating material should be selected taking into consideration the above conditions, molding cycle, and the like.

[0041] As preferable examples of materials which can be coated on the mold cavity surface, polyester resins such as polyethylene terephthalate (45 mN/m), epoxy resins (38 ~ 45 mN/m) and polyimide resins (38 ~ 42mN) and composites of above resins with ceramics, metals, metal oxides and the like, can be mentioned. Among those, polyimide resins or polyimide resin-type composite materials are especially preferable. The surface tension of the polyimide resin at 20 °C is around 38 ~ 42 mN/m when the resin does not contain fluorine atoms and the like, which is perfectly

within the specified range.

[0042] Also, the polyimide resin is especially useful when the surface of a mold having a complicated shape, such as a mold for injection molding, is to be coated. The material to be coated on the surface of the mold is preferably strongly adhered to the surface of the mold so that it will withstand multiple molding cycles. Therefore, the mold surface is preferably coated with a durable film which exhibits strong adhesion to the mold surface for an extended period of time. To form a polyimide resin film on a mold surface having a complicated shape with a strong adhesion between the mold surface and the polyimide film, most preferably, a solution of a polyamide acid in a solvent such as N-methylpyrrolidone, which is a precursor of a polyimide, is coated on the mold surface and heated to form a polyimide resin film. The polyimide precursor polymer exhibits strong adhesion to the mold because of the presence of polar group such as carboxyl group in the precursor. By heat-reacting the polyimide precursor on the surface of the mold surface, the polyimide resin film is formed with strong adhesion to the mold surface.

[0043] Also, in a process of obtaining a molded article by supplying the heat-plasticized resin composition into a mold cavity and cooling while pressing, the mold surface is subjected to heating and cooling cycles with a temperature difference of exceeding 100 °C in each molding cycle. In general, since a difference in the coefficient of thermal expansion between the material coated on the mold surface and a metal is very large, severe stress generates at the interface between the metal and the material coated

on the mold surface in each molding cycle. To withstand its severe stress for more than ten thousand cycles, the material to be coated on the mold surface is required to possess excellent break strength, elongation at break and strong adhesive strength to the metal. Polyimide resins satisfy the above requirements and are preferably used. The most preferable, a linear, high-molecular-weight and tough polyimide resin which does not contain a compound which may interfere with the adhesion to a metal, such as fluorine atom, is the most preferably used.

[0044] Also, to obtain a heat resistant property, the polyimide resin preferably has a high glass-transition temperature (Tg). Although varying depending on the composition of the components, the linear polyimide resin preferably has a Tg of at least 200 °C, or more preferably at least 230 °C. The polyimide preferably has a thermal conductivity as low as possible, in the range of not higher than 0.002 cal/cm·sec·°C.

[0045] The film thickness of the polyimide resin layer is selected within the range of 0.01 ~ 2 mm. When the film thickness is less than 0.01 mm, the effect of the surface improvement of the molded article cannot be observed. When the thickness exceeds 2 mm, the cooling effect of the mold decreases, resulting in the reduction in the molding efficiency. As the molding temperature is lowered, the film thickness of the polyamide resin layer needs to be increased. The optimum film thickness is selected within the range of 0.01 ~ 2 mm. Also, the optimum film thickness of the polyimide depends on the method of molding. For example, in injection molding, the optimum film thickness is in the range of 0.01 ~ 0.5 mm, or

more preferably 0.03 ~ 0.2 mm. On the other hand, in extrusion-blown molding, the optimum film thickness is in the range of 0.1 ~ 1 mm. When the film thickness of the polyamide resin is not uniform, the term "thickness of the polyimide" means the maximum film thickness.

[0046] Furthermore, the linear high-molecular-weight polyimide to be used as the coating material for the mold surface preferably has a strength and elongation as high as possible. Particularly, the polyimide resin preferably has an elongation at break as high as possible, in the range of at least 10%, or more preferably at least 20%. The measurement of the elongation at break is carried out according to ASTD 638. As preferable examples of the high-molecular-weight polyimide resins as the material to be used on the mold surface, Kapton (trade name, from Toray Co., Tg = 428 °C), Novax (trade name, from Mitsubishi Kasei Co. Tg = 399 °C), Upilex R (trade name, from Ube Kosan Co., Tg = 303 °C), Upilex S (trade name, from Ube Kosan Co., Tg = 359 °C), Larc TPI (trade name, from Mitsui Koatsu Kagaku Co., Tg = 256 °C) and PI2088 (from The Upjohn Co., Tg = 342 °C), PAI (from Amoco Co., Tg = 230 °C) can be mentioned.

[0047] Also, as the molds to be used in manufacture of the molded article of the present invention, not only those which are coated with the material having a thermal conductivity of not higher than 0.01 cal/cm·sec·°C (20 °C) and a surface tension of at least 35 mN/m (20 °C) on their cavity wall surfaces, but also those which are entirely made of the materials having a thermal conductivity of not higher than 0.01 cal/cm·sec·°C (20 °C) and a surface tension of at least 35 mN/m (20 °C)

can be used. As a preferable example, an epoxy resin/aluminum composite material [thermal conductivity 0.0034 cal/cm.sec. $^{\circ}$ C (20 $^{\circ}$ C)], trade name Quinnate NEZ, from Zeon Rise Co., can be mentioned.

[0048] [Example]

The present invention will be explained in more detail using Examples and Control Examples. The manufacturing method of the propylene-type resin composition, molding conditions, and testing methods used in each Example are described below.

Manufacturing method of hydroxyl group-containing polypropylene-type resin (OH-PP)

Composition comprising polypropylene, maleic anhydride, and an organic peroxide as shown in the column (I) of the Table 1, were each melt-kneaded using a twin-screw extruder to obtain maleic anhydride-grafted acid-modified polypropylenes. The amount of maleic anhydride in each acid-modified polypropylene was obtained by re-precipitation of the acid-modified polypropylene from xylene/acetone to remove the unreacted maleic anhydride, vacuum-drying of the precipitated modified polypropylene, dissolving the purified modified polypropylene in hot xylene, and titrating the modified polypropylene solution with a sodium methylate standard solution. Furthermore, the obtained acid-modified polypropylenes were each mixed with the hydroxyl group-containing amine compound in an amount as shown in Table 1 (II) and melt-kneaded using a twin-screw extruder to obtain hydroxyl group-containing polypropylene-type resins (OH-PP1 ~ OH-PP5). Also, the

OH-PP6 was obtained by melt-kneading of a mixture of 5 weight parts of a hydroxyl group-containing polypropylene wax with a hydroxyl group content of 0.9 meq/g (Umex H1210, from Sanyo Kasei Co.) and 95 weight parts of propylene-ethylene block copolymer (PP-4, Polypropylene M8840, from Asahi Kasei Co.) using a twin-screw extruder.

Measurement of the hydroxyl group in the hydroxyl group-containing polypropylene-type resin

Measurement of the hydroxyl group in the hydroxyl group-containing polypropylene-type resin was carried out using an acetic anhydride/pyridine method. Detailed explanation of the method of measurement will be described below:

(1) The hydroxyl group-containing polypropylene-type resin was dissolved in hot xylene and precipitated in acetone to purify the resin. The purified sample was filtered and dried under vacuum at 80 °C for at least 8 hours to remove the solvent.

(2) The purified sample in an amount of 3 g was dissolved in 150 ml of a heated mixed solvent of 2:1 (volume ratio) xylene:pyridine and titrated with a 0.05 N Na methylate standard solution using an ethanol solution of phenolphthalein as an indicator to obtain the acid content in 1 g of purified sample.

(3) The purified sample in an amount of 3 g was dissolved in 150 ml of a heated mixed solvent of 2:1 (volume ratio) xylene:pyridine and was mixed with 5 ml of a solution of acetic anhydride in pyridine (1 wt%). The reaction was continued for 2 hours. After the addition of 1 ml of purified

water, the reaction was allowed to proceed for another 2 hours. After the completion of the reaction, the reaction medium was titrated with a 0.05 N Na methylate standard solution using a phenolphthalein solution as an indicator. In the other experiment, the blank testing was carried out. Based on the above procedure, the consumption of the acetic anhydride was obtained.

(4) From the acid equivalent in the purified sample and the consumption of the acetic anhydride, the content of hydroxyl group in the hydroxyl group-containing polypropylene-type resin was calculated. The content of the hydroxyl group was expressed as the equivalent of sodium methylate in 1 g of the hydroxyl group-containing polypropylene-type resin [meq(CH₃ONa)/g(PP)].

Preparation of the test piece for evaluation of the adhesion of the coating film

The test piece for evaluation of the adhesion of the coating film was prepared according to the 3 methods as shown below.

(1) Injection molding method 1

Using an injection-molding machine (Autoshot 50B, from Fanuc Co.) under the conditions of the cylinder temperature of 230 °C and the mold temperature of 60 ~ 120 °C, the molten resin was injected to a chrome-plated mold [thermal conductivity 0.25 cal/cm·s·°C (20 °C)] to obtain a 100 mm x 100 mm x 2 mm flat molded article.

(2) Injection molding method 2

Using an injection-molding machine (Autoshot 50B, from Fanuc Co.)

under the conditions of the cylinder temperature of 230 °C and the mold temperature of 30 ~ 120 °C, the molten resin was injected into a cavity whose surface was coated with a polyimide with a film thickness of 50 μm [trade name Kapton, from Toray Co., thermal conductivity 2.5×10^{-4} cal/cm·s·°C (20 °C), surface tension 38 mN/m (20 °C)] to obtain a flat molded article.

(3) Melt pressing method 1

The molten resin was hot-pressed at 200 °C between PET films [trade name Lumirror, from Toray Co., thermal conductivity 3.6×10^{-4} cal/cm·s·°C (20 °C), surface tension 45 mN/m (20 °C)] to obtain a 100 mm x 100 mm x 2 mm flat molded article.

(4) Melt pressing method 2

The molten resin was hot-pressed at 200 °C between teflon films [Naflon, trade name, from Nichias Co., thermal conductivity 6.0×10^{-4} cal/cm·s·°C (20 °C), surface tension 24 mN/m (20 °C)] to obtain a 100 mm x 100 mm x 2 mm flat molded article.

Measurement of the contact angle of the water droplet

After the molded article surface was cleaned with ethanol, residual ethanol on the surface was wiped off, then the molded article was allowed to stand at 23 °C for 1 hour for drying. A droplet of distilled water with a diameter of 1.5 ~ 2.0 mm was slowly applied onto the molded article surface using a syringe at 23 °C and the humidity of 50 ~ 60%. Ten minutes after the application, the contact angle of the droplet was measured using contact angle measurement equipment (CA-S150 type, from Kyowa Kaimen Kagaku Co.).

Adhesion testing of urethane-type coating

(1) Pretreatment, baking condition

The surface of the molded flat panel with a thickness of 2 mm was wiped out with an ethanol-impregnated cloth. The panel was allowed to dry at 20 ~ 25 °C for at least 1 hour and coated with a 2 component-type urethane coating (Retan PG-60, from Kansai Paint Co.) (film thickness 30 ~ 40 μm), allowed to stand at 20 ~ 25 °C for 30 minutes, and baked at 80 °C for 1 hour in a hot air convection oven. After allowed to stand for 24 hours, the evaluation of the coating film adhesion was carried out following the coating film peeling test as described below.

(2) Coating film peeling test

After the coated test piece was immersed in 40 °C warm water for 240 hours, the surface of the test piece was cut with a cutter knife in such a way that 2 sets of 11 cuts with a spacing of 1 mm were made perpendicular to each other, thus making a lattice of 100 squares. A cellophane adhesive tape (JIS-Z1522) was sufficiently pressed over the above lattice and the tape was peeled off at once at an angle of around 30 degree from the coating surface. The condition of the surface within the lattice was observed. The results were shown as the number of residual coated squares/100.

[0049] [Example 1 ~ 3]

A mixture of 70 weight parts of hydroxyl group-containing polypropylene-type resin (OH-PP1), 30 weight parts of polyolefin-type elastomer (EPR1), and 0.1 weight parts of organic tin compound (DBTL) was melt-kneaded using a twin-screw extruder to obtain a propylene-type resin composition. The obtained resin composition was formed into a molded article according to the molding conditions as shown in Table 2. The contact angle of the water droplet and the adhesion of the urethane-type coating film on the obtained molded articles were evaluated and the results are shown in the lower part of Table 2. The molded articles having a contact angle of the water droplet of 80° or higher exhibited easy peeling of the coated film.

[0050] [Control Examples 1, 2]

The propylene-type resin composition used in Examples 1 ~ 3 was molded according to the molding conditions as described in Table 2. The contact angle of the water droplet and the adhesive strength of the urethane-type coating film on the molded articles were evaluated. The results are shown in the lower part of Table 2. The molded articles having a contact angle of water of 80° or higher exhibited easy peeling of the coated film.

[0051] [Example 4 ~ 10]

The compositions comprising a hydroxyl group-containing polypropylene-type resin (a), a polyolefin-type elastomer (b), an organic tin compound and/or tertiary amine compound (c), other thermoplastic resin and talc in amounts as shown in Table 3 were each mixed and melt-kneaded

using a twin-screw extruder to obtain propylene-type resin compositions. The obtained resin compositions were each molded using the injection molding method 2 to obtain flat molded articles. The contact angle of the water droplet and the adhesion of the urethane-type coating film on the molded articles were evaluated. The molded articles exhibited excellent adhesion of the coated film.

[0052] [Control Examples 3 ~ 5]

Propylene-type resin compositions used in Examples 4, 5, and 6 were molded using the injection molding method 1 to obtain test pieces for evaluation of the coating film. The contact angle of the water droplet and the adhesion of the urethane-type coating film on the molded articles were evaluated. The results are shown in the lower part of Table 3.

[0053] [Table 1]

Hydroxyl group-containing polypropylene-type resin			OH- PP1	OH- PP2	OH- PP3	OH- PP4	OH- PP5	OH- PP6
(I) acid modified polypropylene	polypropylene	PP-1	100					
		PP-2		100		100	100	
		PP-3			100			
	Maleic anhydride		1.0	1.0	1.0	1.0	1.5	
	peroxide		0.1	0.1	0.1	0.1	0.2	
	Degree of addition of maleic anhydride (wt%)			0.45	0.56	0.60	0.56	1.21
	(II) Amino alcohol	Amino Alcohol 1	1.0	1.25	1.25		1.5	
		Amino Alcohol 2				1.2		
	polypropylene	PP-4						95
	Hydroxyl group-containing PP wax							5
	Contents of hydroxyl group (10^{-2} meq/g)		2.20	2.50	2.80	2.10	4.21	1.70

(note) In the above Table, the unit of the numbers is weight parts unless indicated otherwise.

[0054] [Table 2]

	Example 1	Example 2	Example 3	Control 1	Control 2
Molding condition	Molding method	Melt press method 1	Injection molding method 1	Injection molding method 2	Injection molding method 1
	Mold base material	PET	Cr plated	polyimide	Cr plated
	Temperature of base material (°C)	200	110	40	60
Contact angle of water droplet (°, 23°C)	65	75	72	82	90
Adhesion of coating film (no. of residual coated squares/100)	100/100	100/100	100/100	10/100	0/100

The resin composition used:

OH-PP1/EPR1/DBTDL = 70/30/0.1 (weight parts)

[0055] [Table 3]

			Example 4	Example 5	Example 6	Example 7	Example 8
(a) Hydroxyl group-containing polypropylene resin	OH-PP2	90	70		70		
	OH-PP3			60			
	OH-PP4					50	
	OH-PP5						
	OH-PP6						
(b) Olefinic elastomer	EPR-1	10	30		30	50	
	EPR-2			40			
(c) Organic tin and/or tertiary amine compd.	DBTDL	0.1	0.1		0.1	0.2	
	DABCO			0.1	0.1		
Other thermoplastic resin	HDPE						
	LDPE						
	SEBS					15	
Reinforcing agent	talc				15		
Molding condition	Molding method	Injection method 2					
	Temperature of base material (°C)	60	40	80	60	30	
Contact angle of water droplet (°, 23 °C)	72	70	66	72	72		
Adhesion of coating film (no. of residual coated square/100)	100/100	100/100	100/100	100/100	100/100	100/100	

[Table 3 (continued)]

			Example 9	Example 10	Control 3	Control 4	Control 5
Propylene-type resin composition	(a) Hydroxyl group-containing polypropylene resin	OH-PP2			90	70	
		OH-PP3					60
		OH-PP4					
		OH-PP5	30				
		OH-PP6		80			
	(b) Olefinic elastomer	EPR-1	70	20	10	30	
		EPR-2					40
	(c) Organic tin and/or tertiary amine compd.	DBTDL	0.2	0.2	0.1	0.1	
		DABCO					0.1
	Other thermoplastic resin	HDPE	10				
		LDPE		15			
		SEBS					
	Reinforcing agent	talc	10				
Molding condition	Molding method	Injection method 2	Injection method 2	Injection method 1	Injection method 1	Injection method 1	
	Temperature of base material (°C)	60	90	60	80	40	
Contact angle of water droplet (°, 23 °C)		68	74	82	83	84	
Adhesion of coating film (no. of residual coated square/100)		100/100	100/100	0/100	10/100	20/100	

(note) In the above Table, the unit of numbers is weight parts unless indicated otherwise.

[0056] In the above Tables 1 ~ 3, abbreviations are as follows.

Polypropylene

- Homopolypropylene (PP-1)

Polypropylene E 1100 from Asahi Kasei Co.

MFR: 0.5 g/10 min (ASTM D1238)

- Propylene-ethylene block copolymer (PP-2)

Polypropylene M 7100 from Asahi Kasei Co.

MFR: 0.5 g/10 min (ASTM 1238)

- Propylene-ethylene random copolymer (PP-3)

Polypropylene E 3100 from Asahi Kasei Co.

MFR: 0.5 g/10 min (ASTM 1238)

- Propylene-ethylene block copolymer (PP-4)

Polypropylene M 8840 from Asahi Kasei Co.

MFR: 40 g/10 min (ASTM 1238)

Organic peroxides (peroxides)

- 2,5-dimethyl-2,5-di-(t-butylperoxy)hexane

Maleic anhydride

- Crystal Man from Nippon Yushi

Hydroxyl group-containing amine compound

- 2-aminoethanol (aminoalcohol-1)

- 2-(2-aminoethoxy)ethanol (aminoalcohol-2)

Hydroxyl group-containing polypropylene wax

- Umex H 1210 from Sanyo Kasei

Number-average molecular weight: 4,000, hydroxyl number: 50 mg KOH/g

Olefinic elastomer

- Ethylene-propylene copolymer rubber (EPR-1)

Vistalon 805, from Exxon Chem. Co., Mooney viscosity ML_{1+4} (100 °C): 53,
ethylene content: 77 weight%

- Ethylene-propylene copolymer rubber (EPR-2)

Vistalon 878, from Exxon Chem. Co., Mooney viscosity ML_{1+4} (100 °C): 76,
ethylene content: 54 weight%

Organic tin compound

- Dibutyltin dilaurate (DBTDL)

Tertiary ammine compound

- 1,4-diazabicyclo[2.2.2]octane (DABCO)

Other thermoplastic resins

- High-density polyethylene (HDPE)

Santec-HD J 340 from Asahi Kasei, MFR: 7 g/10 min (ASTM D 1238)

- Low-density polyethylene (LDPE)

Santec-LD M 6520 from Asahi Kasei, MFR: 20 g/10 min (ASTM D 1238)

- Hydrogenated Styrene-butadiene-styrene copolymer (SEBS)

Taftec H 1052 from Asahi Kasei, MFR: 12 g/10 min (ASTM D 1238), styrene content: 19 weight%

Reinforcing agent (talc)

- Microace p4 from Nippon talc

[0057] [Effect of the Invention]

The molded article from the propylene-type resin composition of the present invention exhibits excellent adhesion to a urethane-type coating.

The urethane-type coating is generally a coating which forms a coating film by the reaction of a polyisocyanate and a polyol compound and has a one component type and a two component type. Also, a powder coating using a blocked isocyanate is available. The molded article from the propylene-type resin composition of the present invention exhibits excellent adhesion to any of these urethane coatings. Furthermore, since the molded article obtained from the present invention has a smooth surface,

it exhibits excellent appearance after being coated. Also, since the mold to be used in the manufacture of the molded article of the present invention can be used for conventional molding methods, such as injection molding, compression molding, extrusion molding (sheet molding, blow molding) and the like, molded articles with different shapes can be easily obtained and a variety of practically useful articles can be manufactured according to the present invention.